Velocity dependent interactions and a new sum rule in bcc He

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Recent neutron scattering experiments [PRL,**88**,p.195301 (2002)] on solid ^{4}He, discovered a new optic-like mode in the bcc phase. This excitation was predicted by a recently proposed model that describes the correlated atomic zero-point motion in bcc Helium in terms of dynamic electric dipole moments. Modulations of the relative phase of these dipoles between different atoms describes the anomalously soft $T_1(110)$ phonon and two new optic-like modes, one of which was recently found in the neutron scattering experiments. In this work we show that the correlated dipolar interactions can be written as a velocity dependent interaction. This then results in a modified f-sum rule for the $T_1(110)$ phonon, in good agreement with the recent experimental data.

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The phonons in a quantum solid, such as solid He, show various quantum effects arising from the unusually large zero-point motion of the atoms. The strong quantum pressure in solid He lowers the density and introduces large anharmonic terms in the interatomic potential. In bcc He the potential even has a double-well feature [1, 2]. Describing the phonon spectra of this highly anharmonic quantum solid in a consistent manner has therefore presented a theoretical challenge over the years.

The quantum effects appear in the first moment of the dynamic structure factor $S(\omega, k)$, as measured by neutron scattering [3]. The Debye-Waller factor $d(k)$ is unusually small due to the large spread in the atomic positions. In addition there have been measurements [3] of apparent oscillations of the Debye-Waller factor as a function of the scattering wavevector k. These oscillations seemed to be confined to some modes, while absent in the transverse $T_1(110)$ mode. The explanation of these unusual oscillations was given in terms of interference effects between the single and multi-phonon excitations, whereby intensity is transferred between them [1]. The mixing of the phonon modes is attributed to the anharmonicity of the interatomic potential. From symmetry considerations these types of calculations predict that the oscillatory contribution to $d(k)$ vanishes both at the Brillouin zone center and boundary (z.b.).

Recently there has been a new neutron scattering experiment [4] in solid ^{4}He. The main surprise in this experiment was the discovery of a new, gapped excitation branch in bcc ^{4}He. This excitation was found at the energy predicted by a new model of the dynamic effects of the zero-point motion on the phonon spectra [2, 5]. Usually a bcc lattice is not expected to support optic-like phonons, so the measurement of this new excitation branch provides support for the basic idea of the new model, namely the introduction of an additional degree of freedom in bcc He. Within the new model, this degree of freedom is the relative phase of a correlated part of the zero-point motion between the atoms. The latest experiment has also provided data which shows apparent oscillations of the Debye-Waller for the transverse $T_1(110)$ mode [6]. It is this data which we shall analyze in this letter, in terms of the new model.

In previous papers [2, 5] we have presented a new model to describe some of the correlations in the atomic zero-point motion in bcc Helium. The main point is that there is an anharmonic, low energy mode of atomic motion along the major axes of the bcc lattice, which is not described by the harmonic approximation. In bcc He this mode has energy $E_0 \sim 3 - 10$K [2], and is the only free parameter in our model, which we take from empirical data. Additionally, the low mass of the He atom allows relatively large breaking of the Born-Oppenheimer approximation which results in motion-induced electric dipoles, due to relative displacement of the nuclei with respect to the electronic cloud [2, 7]. When these dipoles are uncorrelated they introduce a negligible addition to the usual Van-der Waals interaction. By contrast, correlated and highly directional atomic motion results in dipolar interactions between these dipoles, which is of the order of the energy of the local atomic motion E_0. There can therefore arise a state of quantum resonance between these interacting electric dipoles and the atomic motion. A resonant state between the atomic motion and electric dipolar interactions lowers the ground-state energy of the system. In the ground-state of the bcc phase we found [2] that the dipolar interaction is minimized when the dipoles have the phase relation shown in Fig.1.

The dipolar interactions between the motion-correlated dipoles is [2]

$$E_{dip} \;=\; \sum_{i\neq 0} \mu_0 \cdot \mu_i \left[\frac{3\left(\hat{\mu}\cdot\hat{\mathbf{r}}_{i0}\right)^2 - 1}{\left|\mathbf{r}_{i0}\right|^3} \right] \tag{1}$$

where the electric dipole moment in bcc He turns out to be [2]: $\mu \simeq e \cdot 0.01$, and $\mathbf{r}_{i0} = \mathbf{r}_0 - \mathbf{r}_i$ and the index i runs over all atoms. The resonance condition means that E_0 is the frequency of both the oscillating dipoles and the atomic motion, i.e. $E_{dip} \equiv -E_0/2$ ($\simeq -3$K in bcc ^{4}He). Note that the dipole moments are dynamic, and the expectation value of the dipole moment of the

crystal and of each atom is zero, as required by time reversal symmetry. It is the matrix element of correlated dipoles $\langle \mu_0 \mu_i \rangle$ which is non-zero, and appears in (1).

The modulations of the relative phases of the dipoles on different atoms coincide with only one phonon mode, due to the lower symmetry of the dipolar ground-state compared with the bcc lattice (Fig. 1) [2]. This mode turns out to be the anomalously soft $T_1(110)$ phonon, which agrees very well with the experimental data [2]. In addition our model predicts two new modes which are gapped (optic-like) [2, 5]. One of these modes was recently observed in neutron scattering experiments [4]. Since the dipolar degree of freedom involves atomic motion, it couples to the atomic density operator. A flipping of a dipole with respect to the ground-state configuration (Fig.1) results in a dynamic density fluctuation. A flipping of two adjacent dipoles forms the localized excitation of energy $2E_0$ [2] recently found by neutron scattering [4]. Our model therefore gives a good description of some effects of the anharmonic potential in bcc He, as they are manifested in the excitation spectrum.

Since the dipolar interactions arise from correlated atomic motion, they can be naturally written as a velocity-dependent potential. Such a term will modify the Nozieres-Pines f-sum rule, which results from a Hamiltonian where the momentum operator appears only in the kinetic energy term [8] (i.e. no velocity-dependent interactions). In this paper we calculate the f-sum rule for the $T_1(110)$ phonon, which we describe as a phase modulation of the correlated dipolar array [2].

Before proceeding let us make a short comment concerning the appearance of velocity-dependent interactions in physics. One example is superfluid ^{4}He [9]. In the superfluid a velocity-dependent interaction is introduced into the energy density functional in order to take into account short-range atomic motion correlations. These are termed "backflow" and introduce a non-local velocity-dependent term to the potential energy. The result is a modified f-sum rule for the phonon-roton dispersion in bulk superfluid ^{4}He (Eq.(44) in [9]). All this is very similar to our results, only that we do not introduce any unknown fitting parameters, as is done for the superfluid case.

The dipolar interaction matrix element $X(\mathbf{k})$ in the presence of a phase modulation (i.e. a $T_1(110)$ phonon), is the Fourier transform of (1) [2]

$$X\left(\mathbf{k}\right) = \sum_{i\neq 0} \mu_0 \cdot \mu_i \left[\frac{3\left(\hat{\mu}\cdot\hat{\mathbf{r}}_{i0}\right)^2 - 1}{\left|\mathbf{r}_{i0}\right|^3}\right] \times \exp\left[2\pi i \mathbf{k}\cdot\mathbf{r}_{i0}\right] \quad (2)$$

shown in Fig.2.

By analogy with an oscillating electromagnetic field interacting with an atom [10], we can write the electric dipole moment in term of a local momentum operator

$$\tilde{\mu} = e\tilde{X} \rightarrow \tilde{P}\cdot\frac{e}{m\omega_0} \quad (3)$$

where $\omega_0 = E_0/\hbar$ is the resonance frequency of atomic and dipolar vibration, and the $\tilde{}$ symbol is used for operators.

The space representation of the dipolar interaction energy (1) between the zero-point dipoles in the ground-state can now be written in terms of the local momentum operators

$$\begin{aligned} X &= \sum_{i,j,i\neq j} \tilde{\mu}_j \cdot \tilde{\mu}_i \left[\frac{3\left(\hat{\mu}\cdot\hat{\mathbf{r}}_{ij}\right)^2 - 1}{\left|\mathbf{r}_{ij}\right|^3}\right] \\ &= \left(\frac{e}{m\omega_0}\right)^2 \sum_{i,j,i\neq j} \left[\frac{3\left(\hat{\mu}\cdot\hat{\mathbf{r}}_{ij}\right)^2 - 1}{\left|\mathbf{r}_{ij}\right|^3}\right] \tilde{P}_i\tilde{P}_j \end{aligned} \quad (4)$$

where $\mathbf{r}_{ij} = \mathbf{r}_j - \mathbf{r}_i$. Note that similar to the treatment [9] of superfluid ^{4}He, we find a non-local kinetic term.

The contribution of an interaction X (4) to the f-sum rule is found by performing the usual double commutator with the density operator [8]

$$\begin{aligned} \left\langle\left[\left[\rho_k, X\right], \rho_{-k}\right]\right\rangle &= \frac{\hbar^2k^2}{m}\frac{e^2}{m\omega_0^2} \sum_{i,j,i\neq j} \hat{\mathbf{P}}_j\cdot\hat{\mathbf{P}}_i \left[\frac{3\left(\hat{\mu}\cdot\hat{\mathbf{r}}_{ij}\right)^2 - 1}{\left|\mathbf{r}_{ij}\right|^3}\right] \\ &\quad \times \exp\left[2\pi i\mathbf{k}\cdot\mathbf{r}_{ij}\right] \\ &= N\frac{\hbar^2k^2}{m}\frac{|X(k)|}{E_0} \end{aligned} \quad (5)$$

where $\rho_k = \sum_i e^{-ik\cdot r_i}$ is the standard density operator and $\hat{\mathbf{P}}$ is a unit vector of the local momentum so that we keep track of the phase relation between the correlated motion of different atoms. The commutator (5) is performed at equal times [11]. When calculating the summation in (5) we have to specify the phase relation between the dipoles on different sites. If there is no correlation between the dipoles, the interaction $X(k)$ is zero, and the correlation function of the atomic displacements described by these dipoles, i.e. $S(\omega, k)$, is equally zero. In the bcc phase we have the dipoles correlated in the ground-state as shown in Fig.1, with resulting non-zero dipolar interactions (Fig.2). The momentum operators in (4) replace the usual kinetic energy term for the part of the atomic motion which is described by the dipolar degree of freedom.

Using this double commutator (5) in the usual definition of the dynamic structure-factor $S(\omega, k)$ [8, 12], we get that the new sum rule for the $T_1(110)$ phonon is

$$M_1 = \int_0^\infty d\omega\omega S(\omega,k) = N\frac{\hbar^2k^2}{2m}\left(\frac{2|X(k)|}{E_0}\right)d(k)^2 \quad (6)$$

where just as for usual phonons, the single-phonon contribution to the f-sum rule is modified by the Debye-Waller factor $d(k) = exp^{-\langle u^2\rangle k^2}$ (ACB sum rule)[1, 13], with $\langle u^2\rangle$ being the isotropic zero-point spread of the atomic wavefunction. We assumed here that the dynamic structure factor $S(\omega, k)$, i.e. the correlations in atomic displacements, for the $T_1(110)$ phonon are completely described by the dipolar degree of freedom alone. This is

justified by the excellent agreement between our calculation of the and the $T_1(110)$ spectrum experimental data [2].

The new sum rule we find for the $T_1(110)$ phonon (6) is essentially the usual f-sum rule multiplied by the function $2|X(k)|/E_0$. We find that at $k \to 0$, we recover the usual f-sum rule, since $|X(k \to 0)| \to E_0/2$. While the usual f-sum rule follows from particle conservation, the number of excited dipoles out of the correlated ground-state is not strictly conserved. In particular, when the function $X(k)$ goes to zero at the Brillouin z.b., the commutator (5) vanishes. The vanishing of $M_1(k)$ can be understood as a vanishing of both the restoring force of the dipolar interactions and therefore the dipole-induced correlations between the atomic motion on different sites. Exciting a dipole does not affect the others at this particular momentum, and the neutron can not transfer any energy to this degree of freedom. Note that $M_1 \leq N\hbar^2k^2/2m$ since $2|X(k)| \leq E_0$, so that the energy constraint which specifies that the maximum energy transferred by the neutron to the atom is the given by recoil energy, is satisfied.

Before comparing to the experimental data we note that in the experiments the neutron scattering is usually done in the second or higher Brillouin zone, so the periodic behavior of $X(k)$ (Fig.2) has to be taken into account. The experimental set-up is described elsewhere [3, 4], and both sets of data are for bcc ^{4}He. Our calculation is equally applicable for bcc ^{3}He [7], for which there is at present no neutron scattering data.

The calculated $M_1(k)$ (6) is shown in Fig.3 compared to the neutron scattering data [3, 6]. For $d(k)$ we use the value $\langle u^2 \rangle = 0.17\text{Å}^2$, taken from the measured [14] Debye temperature of $\sim$ 27K. By plotting the ratio $M_1(k)/\left([\mathbf{k}\cdot\mathbf{e}(k)]^2 d(k)^2\right)$ we can easily see deviations from the usual f-sum rule, which gives a constant value ($\mathbf{e}(k)$ being the phonon polarization) [1]. The data in Fig.3 is plotted against the full scattering momentum of the $T_1(100)$ phonon as measured in the most recent experiments [4, 6], so both the older data [3] and the calculation are mapped accordingly.

We find that our calculation is in good agreement with the experimental data. The calculated oscillation in the intensity is observed, in contrast with the usual f-sum rule result. The observed periodicity is different from the one calculated previously for interference terms [1], which gives a correction that vanishes both at the z.b. and the zone center. Additionally, the multi-phonon interference calculation does not predict the complete vanishing of the intensity at any momentum, unlike the result of the model presented here. Note that both our model and previous calculations attribute the oscillations in the scattered intensity to anharmonic effects.

The prediction that the intensity should vanish at the Brillouin z.b. is not observed (Fig.3), only a marked reduction in the scattering intensity. This can arise from the finite resolution of the experiment [4] allowing in stray scattering from other phonons with similar energy (at the z.b. the $T_1(110)$ branch meets other phonon branches). At the z.b. with the smaller momentum ($k \simeq 1.85\text{Å}^{-1}$) there is higher scattering intensity then at the higher momentum z.b. ($k \to 2.85\text{Å}^{-1}$), as expected for stray scattering from other phonons. Future high resolution data at the zone boundaries is needed to resolve this question.

In this work we presented a modified f-sum rule for the anomalously soft $T_1(110)$ phonon mode of bcc He. We find an apparently oscillating Debye-Waller factor and a complete vanishing of the first moment at the Brillioun zone boundary. The new f-sum rule is qualitatively different from the standard response of a harmonic phonon. This unusual result is due to correlations in the atomic zero-point motion, which can be written in terms of a velocity-dependent interaction. The calculated sum rule is then compared with recent neutron scattering data, which does show some of the predicted features. Combined with the recent observation of a new excitation mode [4], the sum rule data presented here provides extra support for the proposed model of correlated zero-point electric dipoles in the ground-state of bcc He [2, 5].

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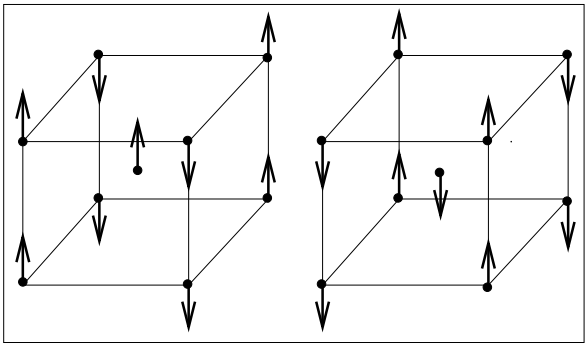

FIG. 1: The two degenerate 'antiferroelectric' dipole arrangements in the ground-state of the bcc phase. The system is in quantum resonance between these two configurations with resonance frequency $\omega_0 = E_0/\hbar$. The arrows show the instantaneous direction of the dipoles along one of the major axes. Similar dipolar arrays exist along the orthogonal major axes.

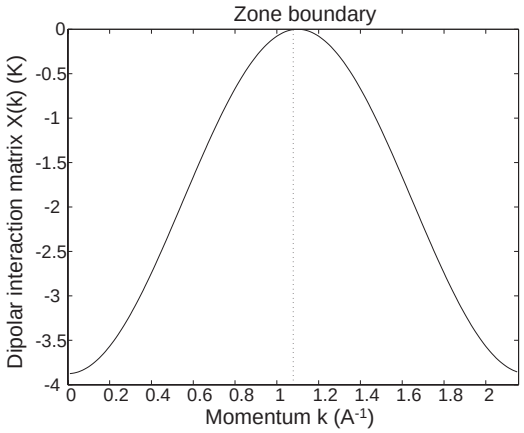

FIG. 2: The dipolar interaction matrix element $X(k)$ (Eq.2) in the bcc phase of ^{4}He, along the (110) direction.

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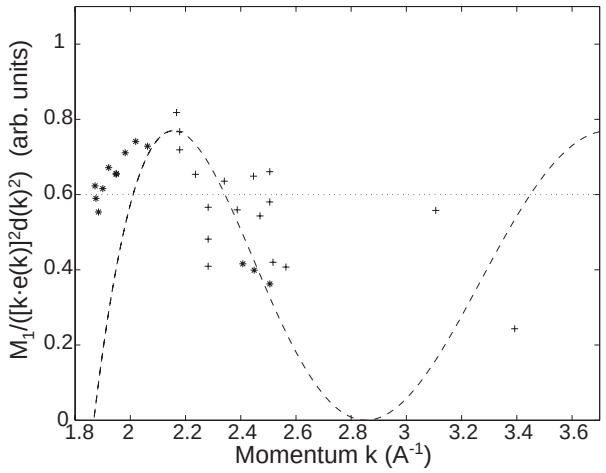

FIG. 3: The ratio $M_1(k)/\left([\mathbf{k}\cdot\mathbf{e}(k)]^2 d(k)^2\right)$ given by Eq.6 (dashed line), compared with the experimental data for the $T_1(110)$ phonon: crosses [3] and stars [6]. The usual form for a harmonic phonon is given here by the horizontal dotted line.